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# Revealing the second harmonic generation in a femtosecond laser-driven cluster-based plasma by analyzing shapes of Ar XVII spectral lines

Eugene Oks,<sup>1,\*</sup> Elisabeth Dalimier,<sup>2</sup> Anatoly Faenov,<sup>3,4</sup> Tatiana Pikuz,<sup>3,4</sup> Yuji Fukuda,<sup>5</sup> Alexander Andreev,<sup>6,9</sup> James Koga,<sup>5</sup> Hironao Sakaki,<sup>5</sup> Hideyuki Kotaki,<sup>5</sup> Alexander Pirozhkov,<sup>5</sup> Yukio Hayashi,<sup>5</sup> Igor Skobelev,<sup>4,7</sup> Sergei Pikuz,<sup>4,7</sup> Tetsuya Kawachi,<sup>5</sup> Masaki Kando,<sup>5</sup> Kiminori Kondo,<sup>5</sup> Alexei Zhidkov,<sup>8</sup> and Ryosuke Kodama<sup>3,8</sup>

<sup>1</sup>Physics Dept., 206 Allison Lab., Auburn University, Auburn, Alabama 36849, USA

<sup>2</sup>LULI-UPMC Univ Paris 06: Sorbonne Universites ; CNRS, Ecole Polytechnique, CEA:Universite Paris-Saclay –F-75252 Paris Cedex 05, France

<sup>3</sup>Institute for Academic Initiatives, Osaka University, Suita, Osaka, 565-0871, Japan

<sup>4</sup>Joint Institute for High Temperatures, Russian Academy of Sciences, ul. Izhorskaya 13/19, Moscow, 125412 Russia

<sup>5</sup>Quantum Beam Science Directorate, Japan Atomic Energy Agency, 619-0215 Kyoto, Japan

<sup>6</sup>Max Born Institute, Berlin 12489, Max-Born str.2a, Berlin, Germany

<sup>7</sup>National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Moscow 115409, Russia

<sup>8</sup>PPC Osaka University and JST, CREST, 2-1, Yamadaoka, Suita, Osaka 565-0871, Japan

<sup>9</sup>ELI-ALPS, Szeged H-6720, Hungary

\*goks@physics.auburn.edu

**Abstract:** We present experiments dealing with a femtosecond laser-driven cluster-based plasma, where by analyzing the nonlinear phenomenon of satellites of spectral lines of Ar XVII, we revealed the nonlinear phenomenon of the generation of the second harmonic of the laser frequency. For performing this analysis we developed new results in the theory of satellites of spectral lines. From such lineshape analysis we found, in particular, that the efficiency of converting the short (40 fs) intense ( $3 \times 10^{18}$  W/cm<sup>2</sup>) incident laser light into the second harmonic was 2%. This result is in the excellent agreement with the 2-Dimensional Particle-In-Cell (2D PIC) simulation that we also performed. There is also an order of magnitude agreement between the thresholds for the SHG found from the line shape analysis and from the 2D PIC simulations.

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**OCIS codes:** (020.2649) Strong field laser physics; (320.2250) Femtosecond phenomena; (190.2620) Harmonic generation and mixing; (350.5400) Plasmas; (020.3690) Line shapes and shifts.

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## 1. Introduction

Various nonlinear phenomena had been observed in optics and plasma physics [1–3] increasing laser intensities and shortening of the pulses, new nonlinear phenomena occur involving many of fields of physics [4–15].

In atomic physics, the interaction of the simplest atoms – hydrogen atoms – with a monochromatic electric field (e.g., a laser field)  $\mathbf{E}(t) = \mathbf{E}_1 \cos(\omega t)$  results in a nonlinear phenomenon in the spectrum of the radiation known as Blochinzew's satellites [16]. Namely, a hydrogen spectral line splits up in a series of satellites at the distances (counted from the unperturbed frequency  $\omega_0$  of the spectral line)  $\Delta\omega = \pm p\omega$ , where  $p = 0, 1, 2, \dots$

As early as in 1933, Blochinzew showed that the intensity  $I_p$  of the satellite at the frequency  $p\omega$  is proportional to the square of the Bessel function  $J_p^2(kE_1/\omega)$ , where  $k = \text{const}$ . At relatively small fields, such as  $kE_1/\omega \ll 1$ , one gets  $J_p^2(kE_1/\omega) = (kE_1/\omega)^{2p}$ , so that practically only the first satellite ( $p = 1$ ) can be observed and its intensity is linear with respect to the energy density of the field ( $I_1$  is proportional to  $E_1^2$ ). However, as the field increases, due to the oscillatory nature of the Bessel functions, the dependence of the satellite intensity

on the energy density of the field becomes highly nonlinear. We note that Blochinzew's result was actually the first example of a more general phenomenon later called "quasienergy spectrum" and "quasienergy harmonics", as introduced by Zeldovich [17] and Ritus [18] in 1964 (Fig. 1(a), 1(b)).

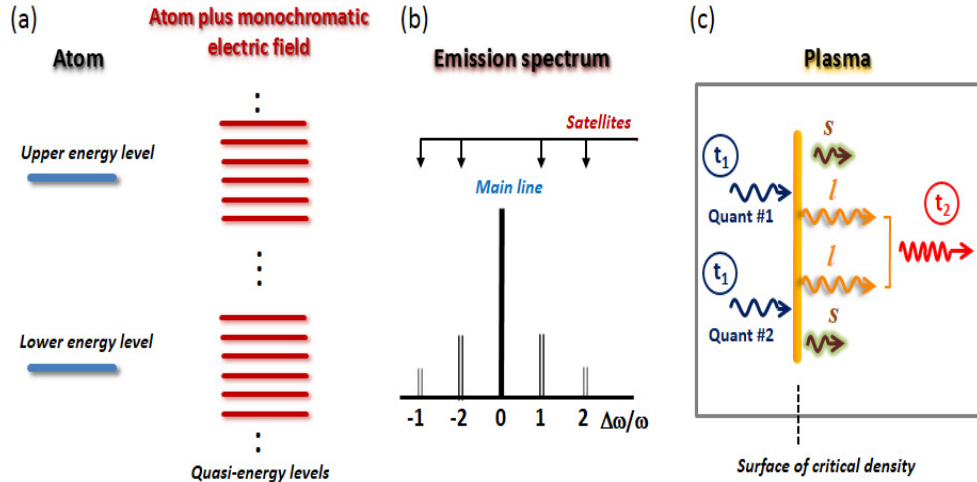


Fig. 1. Schematic of interlinked nonlinear processes. (a) Quasienergy States (QS) of the combined system "atom + electric field  $E(t) = E_0 \cos \omega t$ ". Each atomic/ionic energy level "splits up" into generally infinite number of QS, separated from each other by  $\hbar\omega$ . Only few QS are shown (by red lines). The presence of many other QS (of generally infinite amount) is symbolically indicated by dots, like in the sequence  $\pm \hbar\omega, \pm 2\hbar\omega, \pm 3\hbar\omega, \dots$  (b) The spectrum of radiative transitions between QS shows satellites at  $\pm \omega, \pm 2\omega, \dots$  (in the frequency scale) in addition to the main spectral line. While there is generally an infinite number of QS, practically only a finite number of satellites is observed. (c) Schematic representation of one of the mechanisms of the second harmonic generation – just as an illustrative example. The pump wave ( $t_1$ ) excites both the Langmuir wave ( $l$ ) and the ion-acoustic wave ( $s$ ):  $t_1 \rightarrow l + s$ . Then the predominant process is  $l + l \rightarrow t_2$ , where two Langmuir waves combine to produce a transverse electromagnetic wave ( $t_2$ ) at twice the laser frequency.

In the research area of laser-plasma interactions, one of the important phenomena is Harmonic Generation (HG) – for example, the doubling of the laser frequency [3,6,12,19–24]. Mechanisms of HG involve various nonlinear processes, some of which, in particular, for laser interaction with clusters were considered in [19–24]. For example, in the first step, the transverse laser field ( $t_1$ ) of the frequency  $\omega$  (the pump wave) excites a longitudinal Langmuir wave ( $l$ ). At fairly high laser intensity this occurs via a parametric instability – typically as a parametric decay, which is a nonlinear process where the pump wave ( $t_1$ ) excites both the Langmuir wave ( $l$ ) and an ion-acoustic wave ( $s$ ):

$$t_1 \rightarrow l + s. \quad (1)$$

Parametric instabilities are a common wave-wave interaction. They arise when a nonlinearity such as a pressure gradient couples waves. The physical mechanism is that the pump wave causes oscillations of plasma electrons, which in its turn excite the Langmuir wave and, via the nonlinear coupling – the ion acoustic wave. For this to be possible, the waves must allow frequency and wavenumber matching, which are consequences of energy and momentum conservation. Therefore, the above first step occurs near the surface of the critical electron density

$$N_{ec} = \frac{m_e \omega}{4\pi e^2}, \quad (2)$$

which is the surface where the laser frequency  $\omega$  is equal to the plasma electron frequency  $\omega_{pe}$  of the Langmuir wave.

The second step requires at least one of the two nonlinear processes. The predominant process is

$$l + l \rightarrow t_2, \quad (3)$$

where two Langmuir waves, excited at the first step at the frequency close to the laser frequency, combine to produce a transverse electromagnetic wave ( $t_2$ ) at twice the laser frequency (Fig. 1(c)). The other possible process is

$$l + t_1 \rightarrow t_2, \quad (4)$$

where the Langmuir wave interacts with the laser radiation pump wave to produce a transverse electromagnetic wave ( $t_2$ ) at twice the laser frequency. Both nonlinear processes at the second step involve the Langmuir wave at the frequency  $\omega_{pe}$  close to the laser frequency  $\omega$  – consequently both processes occur near the surface of the critical density. The above is just an illustrative example – there are also other mechanisms of HG [3,6,12,19–24].

The aim of the present work is to develop a novel advanced way of studying an important nonlinear phenomenon in one area of physics by using a different nonlinear phenomenon from another area of physics. Namely, we present experiments dealing with a femtosecond laser-driven cluster-based plasma, where by analyzing the nonlinear phenomenon of (non-Blochinzew) satellites of spectral lines of Ar XVII, we revealed the nonlinear phenomenon of the generation of the second harmonic of the laser frequency in the plasma produced from clusters. This was made possible due to measurements of the x-ray spectra with a high spectral resolution ( $\lambda/\delta\lambda \sim 3000$ ) in a rather broad spectral range under the irradiation of clusters at various laser intensities. This provided insights into the nature of radiation processes of clusters irradiated by ultra-high intensity femtosecond laser pulses. Thus, the nonlinear process of the second harmonic generation manifested via the nonlinear process of the spectral lines satellites.

In Sect. 2 we describe the experiment. In Sect. 3 we analyze the experimental results. In Sect. 4 we complement the analysis by PIC simulations. In Sect. 5 we present conclusions.

## 2. Experiment

The experiments have been performed at Kansai Photon Science Institute, Japan Atomic Energy Agency (KPSI, JAEA, Japan). Two Ti:sapphire laser facilities (wavelength approximately 800 nm) were used. In the first experiment the JLITE-X laser generated 40 fs pulses of the energy of 160 mJ with the contrast of  $10^5$  and the laser intensity up to  $4 \times 10^{17}$  W/cm<sup>2</sup> in vacuum [25]. In the second experiment the J-KAREN laser [26] provided laser pulses with the high contrast of  $10^8$ – $10^{10}$  (with respect to the power; at several picoseconds before the pulse peak. See Fig. 2(d) about the contrast time dependence). The contrast is additionally improved by a saturable absorber inserted between the high-energy CPA oscillator and the stretcher. The ultrafast Pockels cell with the extinction ratio of 200 is applied 500 ps before the main pulse to secure the contrast on the nanosecond time scale. The laser pulse duration was of 40 fs and the pulse energy was of  $\sim 800$  mJ. The intensity of the laser radiation in the focal spot of the diameter of 30  $\mu$ m reached  $3 \times 10^{18}$  W/cm<sup>2</sup> in vacuum [27]. The laser beam was focused about 1.5 mm above the nozzle orifice by an off-axis parabola (Fig. 2).

The Ar clusters were created when a gas of a high initial pressure was expanded into vacuum through a specially designed supersonic nozzle [28–30], which consisted of three

coaxial conical surfaces. The experiments were mainly carried out at the reservoir pressure of 6 MPa of Ar gas, which implies that the mean size of the Ar clusters was  $\sim 1.5 \mu\text{m}$  at the distance of about 1 mm from the edge of the nozzle, where the laser beam was focused (Fig. 2 (a)). With the high contrast of the laser and such a large cluster size, the majority of the Ar clusters were not destroyed by the prepulse until the main laser pulse arrived.

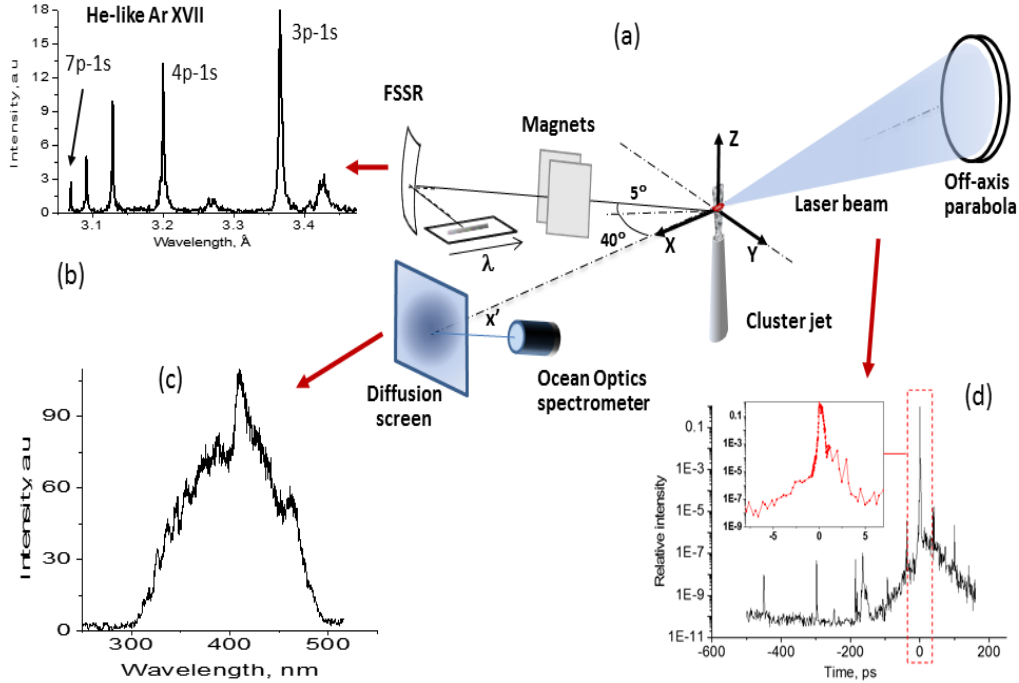


Fig. 2. Schematic experimental setup (a) and a typical X-ray spectrum of Ar cluster plasma in the spectral range 0.305 – 0.35 nm (b), as well as a typical spectrum of the laser beam in the 200 – 500 nm spectral range (c). The laser beam with the contrast shown in (d), ionized Ar clusters and caused nonlinear processes in the cluster plasma. Spectra of Ar XVII were measured by the X-ray spectrometer FSSR. The laser radiation that passed through the cluster plasma was scattered off the screen and spectra of the scattered light in the visible range were measured by the Ocean Optics USB2000 spectrometer.

For high-resolution X-ray spectroscopy measurements a Focusing Spectrometer with Spatial Resolution (FSSR) [31,32] was implemented. The instrument was equipped with a spherically bent quartz (11-20) crystal with the lattice spacing  $2d = 4.912 \text{ \AA}$  and the radius of curvature of  $R = 150 \text{ mm}$ . The crystal was aligned to record K-shell emission spectra of multicharged He-like Ar XVII from the Ly-3 line up to the Ly-7 line in the spectral range of 3.05 – 3.5  $\text{\AA}$ . The FSSR spectral resolving power was approximately 3000. The X-ray spectra from Ar-cluster plasma were observed at the angle of  $\sim 40^\circ$  to the direction of laser beam propagation. The spherically bent crystal was placed at a distance of 500 mm from the plasma source and was centered at wavelength of 3.25  $\text{\AA}$ , which corresponds to the Bragg angle of  $41.5^\circ$ . Spectra were recorded on the X-ray Andor DX-440 back-illuminated charge-coupled device (CCD) with the pixel size  $13.5 \mu\text{m}$ , placed at the distance of 537.7 mm from the source. In such configuration, the spatial resolution along the beam propagation was  $\sim 70 \mu\text{m}$ . The X-ray CCD was protected against the exposure to the visible light using two layers of 1  $\mu\text{m}$  thick polypropylene coated on both sides with  $0.2 \mu\text{m}$  Al. The background fogging and crystal fluorescence due to the intense fast electrons were limited using a pair of 0.5 T neodymium-iron-boron permanent magnets that formed a slit of  $\sim 15 \text{ mm}$  wide in front of the

crystal (Fig. 2). Using such a magnet system allowed us to drastically reduce the level of the background noise in our experiments.

The rigorous noise level analysis was done in the following way. Obtained raw data was averaged by 5 pixels along the spectral resolution using the standard software. As the spectral resolution of our spectrometer was  $\sim 0.19$  mÅ/pixel we estimated that after such a procedure our spectral resolution was  $\sim 1$  mÅ. To reduce additionally the background noise, the spectra presented in Fig. 3 were spatially-averaged over the whole volume of the plasma emission. In distinction, spectra presented in Fig. 4 were averaged only by 4 pixels, which allowed us to have spectra with the spatial resolution corresponding to  $280 \mu\text{m}$ . (We note that the spectra presented in Fig. 5 were also spatially-averaged over the whole volume of the plasma emission for reducing the background noise.) After such procedures the level of noise was rather low, as could be seen from Figs. 3-5, and its effect on the analyzed spectra was not significant.

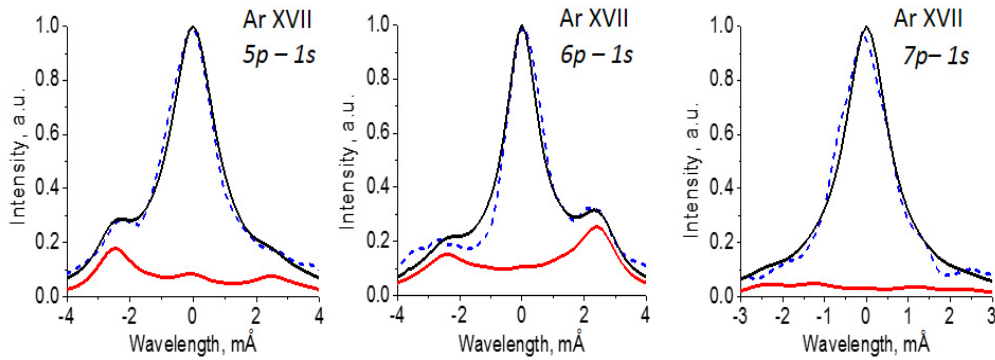


Fig. 3. Comparison of experimental and theoretical spectra of the lines Ar XVII Ly-delta (Ly-5), Ly-epsilon (Ly-6), and Ly-zeta (Ly-7), obtained at laser intensity  $\sim 3 \times 10^{18}$  W/cm<sup>2</sup> (J-KAREN laser facility). Dashed blue – experimental spectra, solid red at the bottom – theoretical spectra from the *bi-chromatic* field region (the field is  $E_1 \cos(\omega t) + E_2 \cos(2\omega t)$ ,  $E_1 = 14$  GV/cm,  $E_2 = 7$  GV/cm), solid black – total theoretical spectra, including the region of no bi-chromatic field.

The combination of the high luminosity of the FSSR spectrometer with the highly sensitive X-ray CCD allowed measuring the spectrum in each laser shot. A typical part of the obtained spectra is presented in Fig. 2 (for a better contrast, these spectra are the summation of 2 shots). For comparison to the experimental data, a forward processing of the calculated spectra was performed including the filter absorption and the CCD response. Additionally, a polypropylene filter of  $6 \mu\text{m}$  thickness was used to reduce the noise level.

Spectral properties of laser beam scattered by the cluster plasma media were studied in the forward direction. We set for this a polytetrafluoroethylene (PTFE) screen at the distance of 63 cm from the plasma in the direction of the laser beam propagation (See Fig. 2(a)). Such a screen allowed measuring the scattered laser radiation in the angle covering  $\sim + -13$  degrees. Spectra of the scattered laser radiation were measured by the OceanOptics USB2000 spectrometer operated in the spectral range 200 – 500 nm without a fiber outside the vacuum chamber. It measured the radiation scattered off the PTFE screen at  $\sim 60$  degrees, which passed through a window and was directed to the spectrometer by an Aluminum mirror. The total distance from the screen to the spectrometer was  $\sim 1.2$  m. To avoid a stray light caused by the laser, the scattered radiation passed through a dichroic beam splitter reflecting out the laser light. A typical scattered laser beam spectrum is presented in Fig. 2(c) and it is clearly seen that the  $2\omega$  laser light dominated it.

It should be noted that observed plasma emission spectra contained not only resonance series of He-like Argon but also their dielectronic satellites. The observation of such



transitions, generally speaking, gives an additional possibility to diagnose plasmas (see, e.g., Rosmej et al [33–36]). However, because we observed time-integrated spectra, we can obtain by this method only time-averaged plasma parameters. For effects considered in the present paper only plasma parameters corresponding to the heating time interval would be relevant, but obtaining them would require a time-resolved X-ray plasma emission. Indeed, the radiation time of resonance lines of He-like Ar lines and their dielectronic satellites is about several ps (see, Sherill et al. [37]), but the laser pulse duration, responsible for the generation of the second harmonic, is 65 fs. Unfortunately, currently it is impossible to measure simultaneously  $2\omega$  “laser” satellites and dielectronic satellites (emitted for much longer time than the  $2\omega$  “laser” satellites) with tens femtosecond time resolution.

### 3. Experimental results and their analysis

Experimental spatially-integrated spectra of the lines Ar XVII Ly-delta (Ly-5), Ly-epsilon (Ly-6), and Ly-zeta (Ly-7), obtained at the laser intensity  $3 \times 10^{18}$  W/cm<sup>2</sup> (the pulse duration 40 fs) are presented in Fig. 3. Experimental profiles of the lines Ly-5 and especially Ly-6 exhibit satellites separated from the line center by  $2\omega[\lambda_n^2/(2\pi c)]$ , where  $\omega$  is the laser frequency and  $\lambda_n$  is the unperturbed wavelength of the Ly-n line of Ar XVII. In other words, the experimental spectra show satellites at the frequencies  $\pm 2\omega$  (counted from the line center in the frequency scale), but do not show satellites at the frequencies  $\pm \omega$ . If the emission would be only from the plasma region where only the wave  $t_2$  exists, it would be fairly easy to interpret the experimental spectra. But this scenario could occur only if the emission originates from the overdense plasma at the density  $N_e$  such that

$$N_{ec} < N_e < 4N_{ec}, \quad (5)$$

which for the laser frequency  $\omega = 2.4 \times 10^{15}$  s<sup>-1</sup> translates into the interval  $1.8 \times 10^{21}$  cm<sup>-3</sup> <  $N_e$  <  $7.2 \times 10^{21}$  cm<sup>-3</sup>. For clarity: in large clusters the laser radiation pump wave  $t_1$  cannot penetrate the plasma of the density higher than  $N_{ec}$ , while its second harmonic  $t_2$  can – but only as long as the density does not exceed  $4N_{ec}$ . We also note that for sufficiently high laser intensity  $I$ , due to relativistic effects, the so-called “relativistic” critical electron density  $N_{ecr}$  might become higher than  $N_{ec}$  [38–40]. For the linearly-polarized laser radiation, according to paper [41],  $N_{ecr} = (\pi a/4) N_{ec}$ , where  $a = \lambda(\mu\text{m}) [I(\text{W/cm}^2)/(1.37 \times 10^{18})]^{1/2}$ . At  $I = 3 \times 10^{18}$  W/cm<sup>2</sup>, the formally-calculated  $N_{ecr}$  does not exceed  $N_{ec}$ , meaning that the threshold for concept of the relativistic critical density to become effective has not been reached.

However, the broadening of the experimental Ar XVII Ly-lines delta, epsilon, and zeta corresponds to noticeably lower densities and thus does not support such scenario. Therefore we have to consider the situation where the observed spectra are formed under the joint action of a relatively strong wave  $t_1$  at the frequency  $\omega$  and a weaker wave  $t_2$  at the frequency  $2\omega$ . (The second harmonic generation produces a wave weaker than the pump wave.) This situation presents quite a challenge for interpreting the experimental spectra. Indeed, it would seem that the stronger wave  $t_1$  at the frequency  $\omega$  should produce relatively strong satellites at the frequencies  $\pm \omega$ ; however, these satellites are not observed.

It is well known that in a high density plasma Rydberg high-n ionic states are not realized due to the Debye screening or the small distance between ions. It is possible to estimate this effect by using hydrogenic wavefunctions for description of Rydberg levels. In this approximation the radius of nl-orbital  $R_{nl}$  can be calculated by the simple formula  $R_{nl} = [3n^2 - l(l+1)]a_0/(2Z)$  ( $a_0$  is the Bohr radius and  $Z$  is the spectroscopic number). For the existence of nl-level it is necessary to satisfy the condition  $R_{nl} < \min\{R_D, R_i\}$ , where  $R_D$  is the Debye radius and  $R_i$  is the distance between ions. It can be seen that even at the electron temperature  $T_e = 100$  eV levels of Ar XVII with  $n < 8$  exist for the electron density up to  $6 \times 10^{21}$  cm<sup>-3</sup>. If the electron temperature is higher, more excited levels with  $n = 8 - 10$  must exist in this density region.

At the electron densities  $\sim 10^{21} \text{ cm}^{-3}$ , Stark broadening of the highly-excited lines Ar XVII Ly-7, Ly-6, and even Ly-5 is practically the same as for hydrogenic systems. This is because the Stark broadening scales as  $\sim n^2$  while the fine structure splitting scales as  $\sim 1/n^3$ . So, the ratio of the Stark broadening to the fine structure scales as  $\sim n^5$ . Therefore there exists a threshold value of  $n$ , starting from which the spectral lines become essentially hydrogenic. In the conditions of our experiments, this threshold value is  $n = 5$ : the Ar XVII lines from  $n = 5, 6, 7$  (and higher) are essentially hydrogenic, while the Ar XVII lines from  $n = 3$  and 4 are not.

While the theory of satellites under the action of a monochromatic field at some frequency  $\omega$  was developed for an isolated lateral Stark component of a hydrogenic line by Blochinzew [16] and for real, multicomponent hydrogenic lines in [42,43], there was no theory of satellites under a bi-chromatic field, such as a relatively strong wave  $t_1$  at the frequency  $\omega$  and a weaker wave  $t_2$  at the frequency  $2\omega$ . Thus, first of all, we had to develop such theory.

Under a *monochromatic* field, the intensity of each satellite is expressed through just one Bessel function, as follows. The spontaneous emission spectrum of a hydrogenic system under a monochromatic field  $E(t) = E_1 \cos(\omega t)$  is controlled by the Fourier expansion of the product of the reduced wave functions

$$\Phi_{n_\alpha \alpha}^* \Phi_{n_\beta \beta} = \exp(ix \mathcal{E}_1 \sin \omega t) = \sum_{p=-\infty}^{\infty} J_p(x \mathcal{E}_1) \exp(ip \omega t), \quad (6)$$

as shown by Blokhinzew [16]. Here  $x$  is the constant of the linear Stark effect, characterizing this Stark component:

$$x = n_\alpha (n_1 - n_2)_\alpha - n_\beta (n_1 - n_2)_\beta, \quad (7)$$

$n_1, n_2$  being the parabolic quantum numbers, and  $n$  being the principal quantum numbers of the upper (subscript  $\alpha$ ) and lower (subscript  $\beta$ ) Stark sublevels involved in the radiative transition;  $\mathcal{E}_1$  is the following dimensionless parameter

$$\mathcal{E}_1 = 3\hbar E_1 / (2Z_{\text{eff}} m_e e \omega), \quad (8)$$

where  $Z_{\text{eff}}$  is the effective charge perceived by the outer electron ( $Z_{\text{eff}} = 17$  for Ar XVII). Therefore the intensity  $I_p$  of the satellite at the frequency  $p\omega$  counted from the unperturbed frequency  $\omega_0$  of the spectral line ( $p = 0, \pm 1, \pm 2, \dots$ ), is

$$I_p = \sum_x j_x \left[ J_{p-2s}(x \mathcal{E}_1) \right]^2, \quad (9)$$

where  $j_x$  are the relative intensities of the Stark components.

In distinction, in a *bi-chromatic* field the intensity  $I_p$  of the satellite at the frequency  $p\omega$  counted from the unperturbed frequency  $\omega_0$  of the spectral line is expressed through a sum of products of two different Bessel functions and the number of terms in the sum is generally infinite, as follows. In a bi-chromatic field  $E(t) = E_1 \cos(\omega t) + E_2 \cos(2\omega t)$ , the right side of Eq. (6) becomes

$$\Phi_{n_\alpha \alpha}^* \Phi_{n_\beta \beta} = \exp[ix(\mathcal{E}_1 \sin \omega t + \mathcal{E}_2 \sin 2\omega t)], \quad (10)$$

where

$$\mathcal{E}_2 = \frac{3\hbar E_2}{2Z_{\text{eff}} m_e e (2\omega)}, \quad (11)$$

This leads to the following expression for the satellite intensity  $I_p$

$$I_p = \sum_x j_x \left[ \sum_{s=-\infty}^{\infty} J_s(x\epsilon_2) J_{p-2s}(x\epsilon_1) \right]^2, \quad (12)$$

which is controlled by the two (rather than one) dimensionless parameters:  $\epsilon_1$  defined by Eq. (8) and  $\epsilon_2$  defined by Eq. (11).

There was also no theory of satellites for *central* Stark components (the components for which  $x = 0$  in Eq. (7)) – whether in a monochromatic or in a bi-chromatic field. The previous theories [16,42,43] in a monochromatic field, as well as the newly developed here theory in a bi-chromatic field, relate to *lateral* Stark components. Ly-n lines with even values of  $n$  have central Stark components. So, we had to develop also a theory of satellites for central Stark components – because at the fairly high laser intensity in the present experiments the satellites of the central Stark component of the Ly-6 line cannot be neglected.

For *central* Stark components ( $x = 0$ ), the instantaneous Stark shift, being quadratic with respect to the instantaneous electric field, is proportional to  $[E_1 \cos(\omega t)]^2 = E_1^2(1 + \cos 2\omega t)/2$ . Therefore, the right side of Eq. (6) becomes

$$\Phi_{n\alpha}^* \Phi_{n\beta} = \exp[i\epsilon_{quadr} \sin 2\omega t], \quad (13)$$

where

$$\epsilon_{quadr} = \frac{n^4(17n^2 - 9m^2 + 19)a_0^3 E_1^2}{64\hbar Z_{eff}^4 \omega}. \quad (14)$$

Here  $m$  is the magnetic quantum number of the Stark sublevels, involved in the emission of the central component, and  $a_0$  is the Bohr radius. The Fourier expansion of the right side of Eq. (13) leads to the following result for the central component contribution  $I_{c,2p}$  to the intensity of the satellite at the frequency  $2p\omega$  counted from the unperturbed frequency  $\omega_0$  of the spectral line ( $p = 0, \pm 1, \pm 2, \dots$ )

$$I_{c,2p} = j_c [J_p(\epsilon_{quadr})]^2, \quad (15)$$

where  $j_c$  is the relative intensity of the central Stark component.

The primary task in interpreting the experimental spectra, based on these newly developed theories, was to find a pair (or pairs) of  $\epsilon_1$  and  $\epsilon_2$  satisfying the following criterion: the intensity  $I_2$  of the 2nd satellite significantly exceeds the intensity  $I_1$  of the 1st satellite. It turned out that if in a particular laser shot the satellites are observed only in one spectral line, then there are several pairs of  $\epsilon_1$  and  $\epsilon_2$  satisfying the criterion, thus making it impossible to select a single pair of  $\epsilon_1$  and  $\epsilon_2$ .

However, if in a particular laser shot the satellites are observed in at least two spectral lines (such as, e.g., Ly-5 and Ly-6 lines in Fig. 3), then it is possible to find a single pair of  $\epsilon_1$  and  $\epsilon_2$  satisfying the above criterion. In particular, for this case the theoretical analysis, through variations of different plasma and laser parameters, led to the following pair:  $\epsilon_1 = 4$ ,  $\epsilon_2 = 1$ . According to Eq. (8), this translates into the electric field amplitudes  $E_1 = 14$  GV/cm,  $E_2 = 7$  GV/cm.

In these shots the laser intensity was  $3 \times 10^{18}$  W/cm<sup>2</sup>, corresponding to the electric field amplitude in vacuum  $E_0 = 48$  GV/cm. Thus, the conversion efficiency, defined as the ratio of the energy density in the generated second harmonic in the plasma to the energy density of the incident laser field in vacuum, was 2%.

For fitting the experimental profiles of the Ly-delta and Ly-epsilon lines it was necessary to take into account that the observed emission is a superposition of the emission from a more dense plasma region, where there are waves  $t_1$  and  $t_2$ , with the emission from a less dense, but a larger plasma region, where there are no periodic electric fields. In this situation the

theoretical spectrum of a line consists of a relatively broad pedestal, emitted from the periodic-field-region, and a relatively narrow, but more intense component, emitted from the no-periodic-field-region.

We mention the fact that while the electron densities of the periodic-field region and of the no-periodic-field region are noticeably different ( $\sim 3 \times 10^{20} \text{ cm}^{-3}$  and  $\sim 1 \times 10^{20} \text{ cm}^{-3}$ , respectively, as found from the best fit to the experimental profiles), the Stark broadening by ion and electron microfields does not differ that much in these two regions as compared to how they would differ if it were not for the following phenomenon. The fairly strong field of the wave  $t_1$  dramatically *suppresses the Stark broadening by the ion microfield* in the periodic-field region. This phenomenon was predicted theoretically in [44] and later confirmed experimentally in a tokamak plasma [45] (see also book [43] Sects. 4.3.1 and 7.5.1). Since for the hydrogenic multicharged systems the predominant mechanism of the Stark broadening is the broadening by the ion microfield, its suppression by the strong field of the wave  $t_1$  leads to a significant narrowing of the Stark profile of each satellite from the periodic-field region. As a result, the Stark width of each satellite does not differ much from the Stark width of the emission from the no-periodic-field region.

We also took into account the theoretically expected asymmetry of the profiles. For the theory of the asymmetry we could refer to papers [46,47] and references therein.

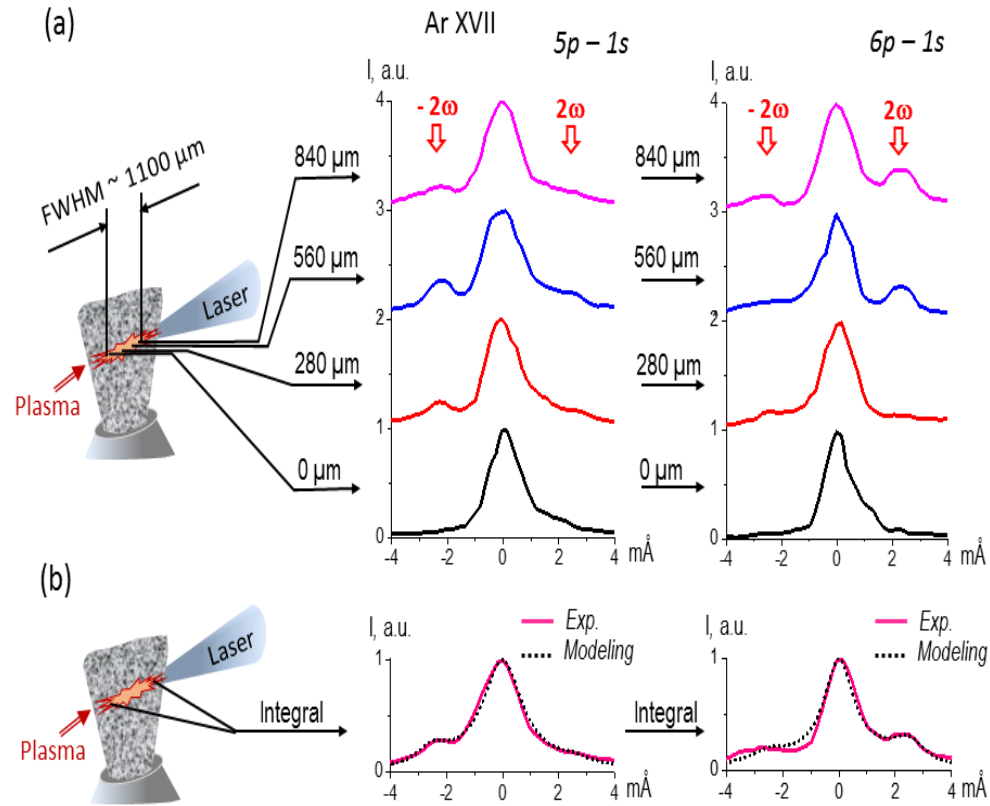


Fig. 4. Experimental and theoretical spectra of the lines Ar XVII Ly-delta (Ly-5) and Ly-epsilon (Ly-6), obtained at laser intensity  $\sim 3 \times 10^{18} \text{ W/cm}^2$ . (a) Spatially ( $\sim 280 \mu\text{m}$ )-resolved experimental spectra along the laser beam propagation. The most pronounced  $2\omega$  satellites are observed close to the center of plasma channel; (b) Spatially-integrated experimental spectra (solid red), for which the modeling (dashed black) was provided. There is a good agreement of the experimental and modeled results.

In addition we clarify that the Doppler and the opacity broadenings were also taken into account. However, the opacity broadening very significantly decreases as the principal quantum number  $n$  increases. Therefore, the ratio of the Stark broadening (which scales as  $\sim n^2$ ) to the opacity broadening decreases very dramatically as  $n$  increases. The Doppler broadening is practically independent of  $n$ , so that the ratio of the Stark broadening to the Doppler broadening scales as  $\sim n^2$  and rapidly increases with  $n$ . For these reasons, the Stark broadening of the lines from the levels  $n = 5, 6$ , and  $7$  predominated over the Doppler and opacity broadenings (though the latter two broadenings were taken into account).

Figure 3 shows the theoretical profiles providing the best fit to the corresponding experimental profiles under laser intensity  $3 \times 10^{18} \text{ W/cm}^2$ . It is seen that the bi-chromatic field, taken into account via our newly developed theory, yields a reliable interpretation of the experimental data. The average root-mean-square deviation of the simulated profiles from the corresponding experimental profiles is less than 5%.

We emphasize that the dominance of the 2nd satellites in the spectrum of just one line could be explained by a monochromatic field. However, the dominance of the  $2\omega$  satellites in the spectra of two lines (Ly-5 and Ly-6) can be explained only by a bi-chromatic field.

For clarity: the “laser” satellites are much more intense for hydrogenic spectral lines compared to non-hydrogenic spectral lines. Therefore, under the conditions of our experiment we did not observe the  $2\omega$  satellites in lines from  $n = 3$  and  $4$ . As for the hydrogenic line from  $n = 7$ , at the possible location of the  $2\omega$  satellites, the intensity of this spectral line declined (compared to the center of the line) to the level close to the level of noise, so that there is zero or little visibility of the  $2\omega$  satellites.

We would like to mention that  $2\omega$  satellites could be clearly seen not only in spatially-integrated spectra presented in Fig. 3, but also in the spectra spatially-resolved ( $\sim 280 \mu\text{m}$  resolution) along the laser beam propagation in the Ar cluster media, as presented in Fig. 4. It shows that  $2\omega$  satellites practically disappear at the entrance of the laser beam in the plasma and at the exit of the laser beam from the plasma, while  $2\omega$  satellites are very pronounced in the central part of the plasma, where the laser beam intensity reaches its maximum. (In Fig. 4 we do not show the spatially resolved spectra of 7p-1s line because its intensity is very low at each spatial “slice” of the plasma and only the spatially-integrated spectrum of this spectral line presents meaningful data, as shown in Fig. 3)

Figure 5 shows (in dashed blue) the experimental spectra of the lines Ar XVII Ly-5 and

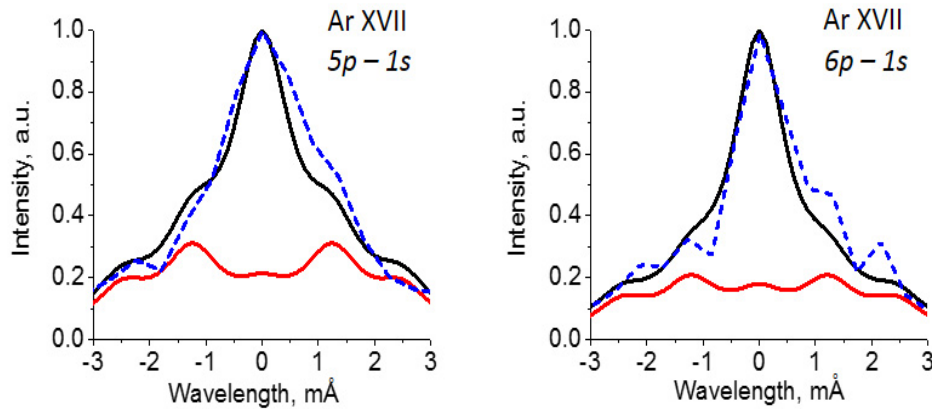


Fig. 5. Comparison of experimental and theoretical spectra of the lines Ar XVII Ly-5 and Ly-6, obtained under laser intensity  $4 \times 10^{17} \text{ W/cm}^2$  and laser pulse duration 500 fs (JLITE-X laser facility), with modeled ones. Dashed blue – experimental spectra, solid red at the bottom – theoretical spectra from the *monochromatic* field region (the field is  $E_1 \cos(\omega t)$ ,  $E_1 = 10.6 \text{ GV/cm}$ ), solid black – total theoretical spectra, including the region of no monochromatic field.

Ly-6 obtained at the laser intensity  $4 \times 10^{17}$  W/cm<sup>2</sup>, which is an order of magnitude lower than the laser intensity discussed in previous Fig. 3. From the theoretical analysis it turned out that these experimental profiles do *not* show a signature of the SHG. These profiles can be fitted by using just the *monochromatic* (not bi-chromatic) field at the laser frequency, as shown in Fig. 5. The average root-mean-square deviation of the simulated profiles from the corresponding experimental profiles is less than 10%.

The above does not mean that at the laser intensity  $4 \times 10^{17}$  W/cm<sup>2</sup>, there was absolutely no SHG. Rather it means that if SHG was present, its amplitude was too small to be detected via spectral line profiles. Thus, it seems that the laser intensity threshold for SHG, deduced from the spectroscopic analysis of above spectra presented in Fig. 5 is about  $4 \times 10^{17}$  W/cm<sup>2</sup> or slightly smaller.

#### 4. PIC simulations

In addition to the parametric processes (discussed in our paper just as examples) generating the second harmonic of the laser radiation, there are other nonlinear mechanisms capable of producing harmonics in laser plasmas at high laser intensity (see, e.g., paper [48]). Because of the complexity of these processes, we performed the simulations of the interaction of a short pulse of the intense linearly-polarized laser radiation with clusters of a large diameter using a 2D Particle in Cell (PIC) code [49,50] under the conditions close to our experiments. It was done by a modified 2D LSP-code [49] employing 30 particles in a cell of the size 4 nm times 4 nm and using a simulation box of the size 20  $\mu$ m times 20  $\mu$ m. The laser intensity was chosen as  $3 \times 10^{18}$  W/cm<sup>2</sup> with the pulse duration of 40 fs and the focal spot size of 4 microns. The Ar<sup>+16</sup> cluster had the diameter of 500 nm and the density of  $10^{22}$  cm<sup>-3</sup>. The results of the simulations are shown in Fig. 6.

One can see in Fig. 6(a) the strong modification (“quasi-spherical” waves) of the initial laser pulse (“plane” waves) at the propagation through the cluster. The scattered light contains harmonics of the incident light. The conversion efficiency into the second harmonic was found to be about 2%, which is in excellent agreement with the result obtained by X-ray spectroscopic measurements and theoretical modeling.

Figure 6(a) shows also that harmonics propagate at a relatively small angle (about 10 degrees)

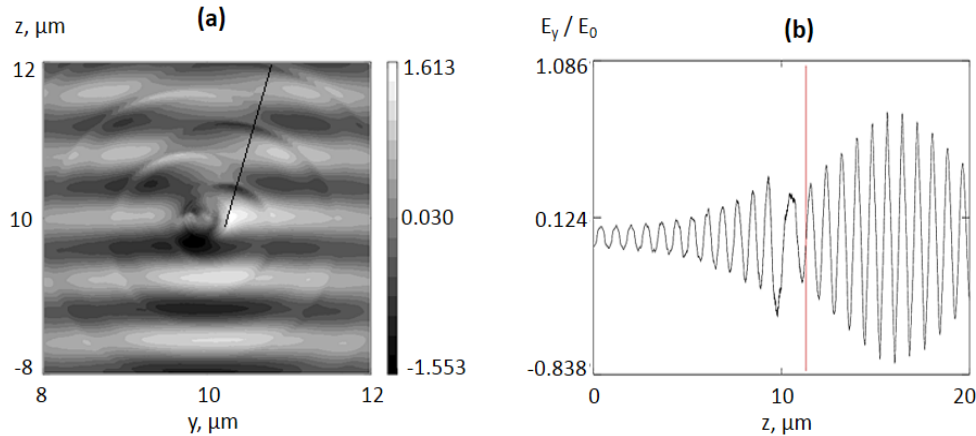


Fig. 6. Simulation of the interaction of a 40 fs pulse of intense ( $3 \times 10^{18}$  W/cm<sup>2</sup>) linearly-polarized laser radiation with the Ar<sup>+16</sup> cluster of the diameter 500 nm using a 2D PIC code. (a) The 2D spatial distribution of electric field  $E_y(y, z)$  at  $t = 42$  fs. The laser pulse propagated from bottom to the top of the figure along the  $z$ -axis. The cluster center is located at the point  $z = y = 10 \mu\text{m}$ . (b) The distribution of electric field  $E_y(y, z)$  for  $y = 10 \mu\text{m}$  at  $t = 64$  fs.

with respect to the direction of the laser beam. This is consistent with the geometry of the experiment presented in Fig. 2.

We considered the area (see red line in Fig. 6(b)) at  $z > 11.4 \mu\text{m}$  to avoid influence of the the ambipolar electric field. It is seen that after the propagation of laser pulse through the cluster, the initial monochromatic field is getting harmonics of the fundamental frequency. The spectrum has only a weak dependence on the position of this line.

Our PIC simulations show that there is a significant decrease of harmonics amplitude  $a_h$  with the increase of the cluster diameter  $r_c$  as  $a_h \sim r_c^{-6}$ . The conversion in harmonics considerably decreases with the increase of the plasma density scale length  $L_n = c_s t_{lp}$  (i.e. contrast) till the value equal to the amplitude of the electron oscillations in a laser field:  $r_E = eE/m\omega^2$ . Here  $c_s[\text{cm/s}] = 10^6 T^{1/2}$  is the ion acoustic velocity, the temperature of cold electrons is  $T[\text{eV}]$ , and the duration of a laser prepulse is  $t_{lp}$ . For the contrast  $10^8$  or the prepulse intensity of  $3 \times 10^{10} \text{ W/cm}^2$  the temperature of cold electrons is about  $T = 10 \text{ eV}$  at the duration of the laser prepulse of  $t_{lp} = 10 \text{ ps}$ . From here one can get  $L_n = 300 \text{ nm}$ , but in the case of the higher contrast  $10^{10}$  this scale is 10 times less. At our laser intensity, we have  $r_E = 150 \text{ nm}$  – therefore, in the case of the first contrast, the harmonics will be suppressed in comparison to the second one if the laser absorption is similar. Of course, for even lower laser contrast  $\sim 10^5$ , no  $2\omega$  harmonics should be produced in cluster media even for clusters of a large size, which is in a good agreement with the spectra presented in Fig. 7(a), where in such case no  $2\omega$  laser satellites were observed.

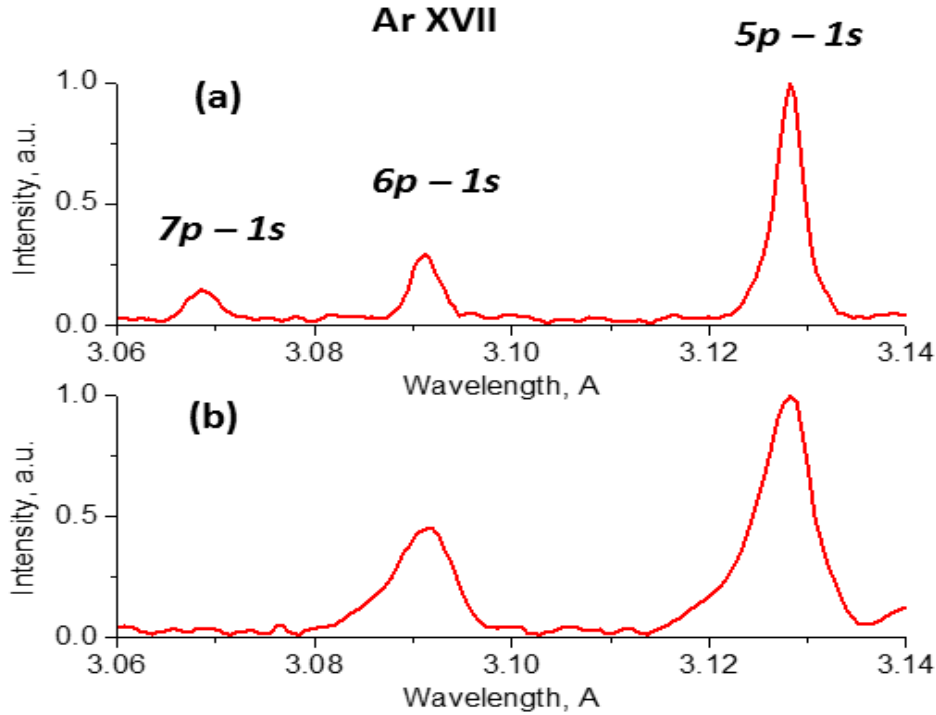


Fig. 7. Experimental spectra of the lines Ar XVII Ly-delta (Ly-5), Ly-epsilon (Ly-6), and Ly-zeta (Ly-7), obtained while decreasing either the laser intensity, or the laser contrast, or the cluster sizes. (a) Laser intensity =  $1.8 \cdot 10^{18} \text{ W/cm}^2$ , pulse duration = 30 fs, contrast =  $10^5$ , cluster size  $> 1 \mu\text{m}^3$ ; (b) Laser intensity =  $1 \cdot 10^{17} \text{ W/cm}^2$ , pulse duration = 45 fs, contrast =  $10^5$ , cluster size  $\sim 70 \text{ nm}$ . It is seen that no  $2\omega$  laser satellites were observed, which is in a good agreement with the PIC simulation results for lower laser intensities, or lower contrasts, or smaller sizes of the clusters.

Our PIC simulations also show that at significantly lower laser intensities there was practically no harmonics generation. The threshold was found to be of the order of  $\sim 10^{17}$  W/cm<sup>2</sup> because this nonlinear process requires a relativistic laser intensity. This PIC simulation result is in a good agreement with the analysis of the experimental spectra observed at lower laser intensities of  $(1-4) \times 10^{17}$  W/cm<sup>2</sup>. Indeed, there are no  $2\omega$  satellites in the experimental spectral lines presented in Fig. 7(b); there was no need to engage the bi-chromatic field for interpreting the experimental spectral lines presented in Fig. 5. The conclusion, based on the analysis of the experimental lineshapes, that there is no second harmonic generation for lower laser intensities, or lower contrasts, or smaller sizes of the clusters, is in a good agreement with the PIC simulation results.

## 5. Conclusions

In the experiments, where the intense femtosecond laser radiation interacted with clusters, we revealed the nonlinear phenomenon of the second harmonic generation (SHG) in the cluster-based plasma using the nonlinear phenomenon of the satellites of spectral lines of Ar XVII. This opens up the way for diagnosing the SHG in laser-produced plasma by analyzing the shapes of the emitted spectral lines.

From such lineshape analysis we found, in particular, that the efficiency of converting the short (40 fs) intense ( $3 \times 10^{18}$  W/cm<sup>2</sup>) incident laser light into the second harmonic was 2%. This result is in the excellent agreement with the 2D PIC simulation that we also performed. There is also a good match of the SHG threshold found from the line shape analysis and from the 2D PIC simulations.

On the experimental side, the novelty of our work is observing the X-ray spectroscopic signature of the second harmonic generation in a cluster-based plasma produced by a femtosecond laser. On the theoretical side, we developed the first ever theory of satellites of lateral Stark components in the bi-chromatic field and of satellites of the central Stark components.

Our results should be important simultaneously for several areas of physics: atomic physics, plasma physics, laser-plasma interactions, and physics of clusters.

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